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730-060, Kyungsangbuk-do (KR). **CHUNG, Jae-Sik** [KR/KR]; 109-601 Sunkyoung Apt., Yatap-dong, Bundang-ku, Seongnam-city 463-070, Kyungki-do (KR).

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(74) Agent: **KIM, Seong-Ki**; 9th Teheran Building, 825-33, Yoksam-dong, Kangnam-ku, Seoul 135-080 (KR).

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(71) Applicant (*for all designated States except US*): **LG CHEMICAL LTD.** [KR/KR]; LG Twin Tower, 20 Yoido-dong, Yongdungpo-ku, Seoul 150-721 (KR).

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(72) Inventors; and

(75) Inventors/Applicants (*for US only*): **KIM, Hyeong-Jin** [KR/KR]; 6-101 LG Apt., 381-4 Doryong-dong, Yousung-ku, Taejeon-city 305-340 (KR). **YOO, Seung-II** [KR/KR]; 432-21, Wonnam-dong, Kumi-city

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**WO 01/52340 A1**

(54) Title: NEW ELECTROLYTES AND LITHIUM ION BATTERY USING THE SAME

(57) Abstract: The present invention relates to a lithium ion battery, more particularly to a new electrolyte and a lithium ion battery which comprises the same using an anode including graphitized carbon and a cathode including lithium-containing transition metal oxide. The present invention provides a compound of 4-carbomethoxymethyl-1,3-dioxan-2-one or 4-carboethoxymethyl-1,3-dioxolan-2-one represented by formula (1) which comprises both a cyclic ring carbonate structure and a linear carbonate structure, a lithium-containing electrolyte which includes the compound of formula (1), and a lithium ion battery which includes the electrolyte using the anode including graphitized carbon and the cathode including lithium-containing transition metal oxide. The lithium ion battery of the present invention fabricated by using the new compound has high electric capacity because of the graphitized carbon, and superior charge-discharge cyclic life characteristic and low temperature performance.

## NEW ELECTROLYTES AND LITHIUM ION BATTERY USING THE SAME

This application claims priority of Korea patent Application No. 2000-000934, filed on January 10, 2000.

5

### **BACKGROUND OF THE INVENTION**

#### **(a) Field of the Invention**

The present invention relates to a lithium ion battery, more particularly to a new electrolyte and a lithium ion battery which comprises the same, using an anode including graphitized carbon and a cathode including  
10 lithium-containing transition metal oxide.

#### **(b) Description of the Related Art**

Ever since the lithium ion liquid secondary battery in various electrochemical batteries had been for the first time commercialized by Sony  
15 Co., the lithium battery has been used increasingly in portable computers and cellular phones etc., instead of the lithium ion secondary batteries of prior art as it has a high energy density. The lithium ion liquid secondary battery comprises an anode including carbonaceous material as an anode active material and a cathode including metal oxide of  $\text{LiCoO}_2$  etc. as a  
20 cathode active material, and was prepared by intercalating a porous polyolefin-based separator between the anode and the cathode, then by injecting a non-aqueous electrolyte having a lithium salt of  $\text{LiPF}_6$  etc. When the battery charges, the lithium ions of the cathode active material are released and then are inserted into the carbon layer of the anode. When  
25 the battery discharges, oppositely the lithium ions of a carbon layer of an anode are released and then are inserted into the cathode active material.

The non-aqueous electrolyte plays a mediating role moving the lithium ions between the anode and the cathode. The electrolyte should be stable within the scope of the operation voltage of the battery, and be able to transfer the ion sufficiently with fast velocity.

5       As an electrolyte, U.S. Pat. Nos. 5,521,027 and 5,525,443 disclose an admixture electrolyte of a linear carbonate and cyclic carbonate. The cyclic carbonate has a large polarity and thus is sufficiently capable of dissociating lithium, but has low ion conductivity due to the large viscosity. Therefore, in these patents, mixing linear carbonate with a low polarity and a  
10       low viscosity reduces the viscosity of the electrolyte comprising the cyclic carbonate.

      The above cyclic carbonate includes carbonate such as ethylene carbonate (EC), propylene carbonate (PC), vinylene carbonate (VC), butylene carbonate (BC) etc. PC has good low temperature performance  
15       due to a low freezing point of  $-49^{\circ}\text{C}$ . However, if an anode uses graphitized carbon of a large capacity, there is the problem of a sudden reaction between PC and an anode when the battery is charging. Thus, EC is commonly used, as it forms the most stable protecting film among a cyclic carbonate in the battery comprising an anode using graphitized carbon.  
20       However, if EC is used in large amount, the low temperature performance of the electrolyte is abruptly deteriorated due to the EC having a high melting point of  $37^{\circ}\text{C}$ . To resolve this problem, it is common use a two-component electrolyte mixing a linear carbonate having a low melting point and a low viscosity as a second component with the EC.

25       The above linear carbonate includes a carbonate such as dimethyl carbonate (DMC), diethyl carbonate (DEC), and ethyl methyl carbonate (EMC) etc. If the EMC having the lowest melting point of  $-55^{\circ}\text{C}$  is used, the battery exhibits an excellent low temperature performance.

However, merely mixing cyclic carbonate and linear carbonate does not satisfy the need for a high capacity and low temperature performance in the lithium ion battery.

In fact, there has been research to improve the low temperature performance of electrolyte comprising the EC, by adding another electrolyte or new additives to the electrolyte of prior art. The literature (J. Electrochem. Soc. 146(2), 485, 1999) discloses that an electrolyte of the three-component system mixing EC, diethyl carbonate (DEC), and dimethyl carbonate (DMC) has an excellent low temperature performance, better than the two-component system. Other literature (J. Fluorine Chem. 87 (1998) 221) discloses that low temperature performance is improved by adding  $\text{CHF}_2\text{COOCH}_3$  to an electrolyte comprising EC and DEC.

If additives are added to the electrolyte like the above methods, it can improve the ion conductivity due to a lower freezing point and lower viscosity at low temperature, as the molecules of the electrolyte are prevented from forming a regular configuration with each other. That is, if the electrolyte of more than the three component wherein a suitable organic material as the third component is further added to the electrolyte of the two-component is used, there is the effect of a freezing point depression when the electrolyte has suitable composition, and effect of improving the charge performance of low temperature due to a reduction of viscosity at low temperature.

In fact, the electrolyte must be shown to be electrochemically stable within the scope of the operation voltage of the battery, and must be shown to have low reactivity with the graphitized carbon, and must not shorten the charge-discharge cyclic life by forming a stable protecting film on an anode. However, there is no an electrolyte which is known to satisfy the above condition.

### SUMMARY OF THE INVENTION

It is an object of the present invention to provide a new compound that is electrochemically stable within the scope of the operation voltage of the battery, as the compound has both a cyclic carbonate structure and a linear carbonate structure in molecule. The compound has a low reactivity with graphitized carbon of high electric capacity, and does not shorten a charge-discharge cyclic life by forming a stable protective film on an anode and thus can be used as a component of electrolyte.

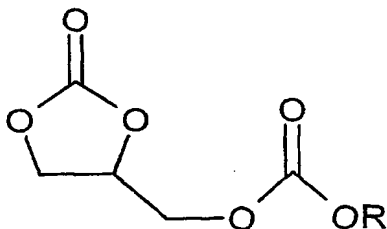
It is other object to provide an electrolyte comprising the above new compound.

It is another object of the present invention to provide a lithium ion battery having a large electric capacity and a superior low-temperature performance comprising an anode including a graphitized carbon and a cathode including a lithium-containing transition metal oxide, a porous separator, and an electrolyte including a lithium salt and the above new compound.

### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

In order to achieve the above objects and others, the present invention provides a compound shown in Formula 1:

[Formula 1]

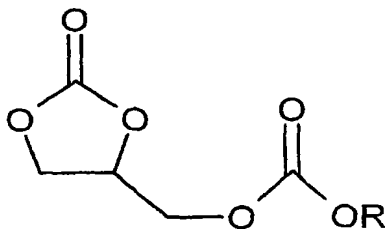


wherein, R is methyl or ethyl group.

The present invention also provides an electrolyte comprising:

- a) a lithium salt; and  
b) the following compound shown in Formula 1 comprising both a cyclic carbonate structure and a linear carbonate structure in molecule:

[Formula 1]



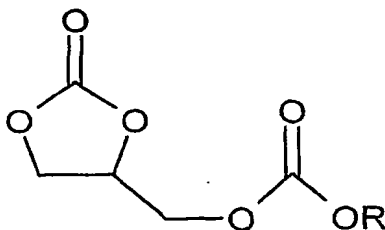
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wherein, R is methyl or ethyl group.

The present invention also provides a lithium ion battery comprising an anode including graphitized carbon as an active material, capable of absorbing and releasing lithium ions reversibly, a cathode including lithium-  
10 contained transition metal oxide as an active material, capable of absorbing and releasing lithium ions reversibly, a porous separator, and an electrolyte, wherein the electrolyte comprising:

- a) a lithium salt; and  
b) the compound shown in Formula 1 comprising both a cyclic  
15 carbonate structure and a linear carbonate structure in molecule:

[Formula 1]



wherein, R is methyl or ethyl group.

20

The present invention is described in detail as follows.

The present invention relates to the compound shown in Formula 1

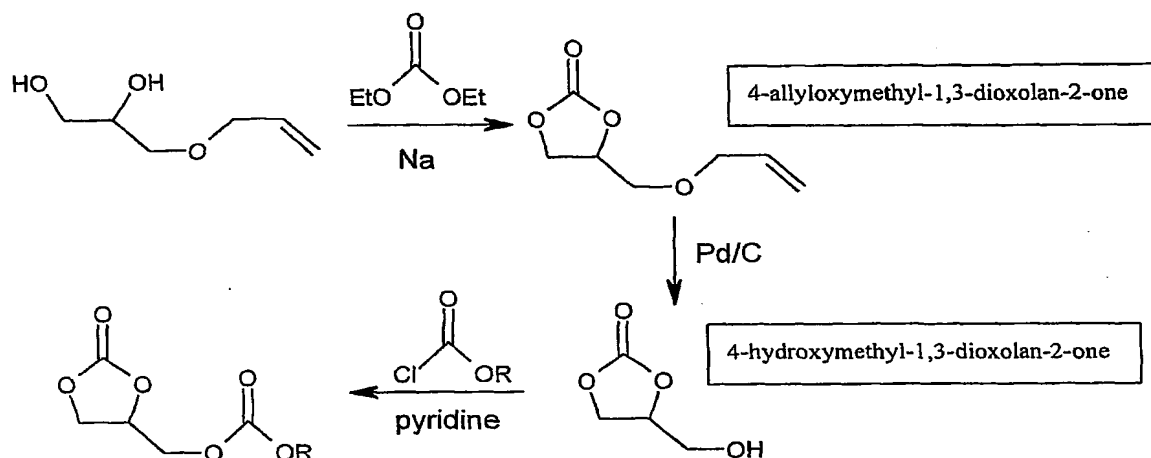
as an additives for an electrolyte of the lithium ion battery comprising an anode including a graphitized carbon, a cathode including a lithium-containing transition metal oxide, a porous separator, and a lithium salt-containing electrolyte.

5           Since the compound comprises both a cyclic carbonate structure and a linear carbonate structure in molecule, the battery can possess a large capacity and improve low temperature performance when the compound is used for the lithium ion the secondary battery including a lithium salt. In particular, the compound is electrochemically stable within scope of the  
10 operation of the battery, has low reactivity to graphitized carbon, and forms a stable protecting film due to a little possibility of insertion of the carbonaceous material of an anode together with the lithium ion, as the molecular size of the compound is relatively larger than those of EC and PC.

          In the compound shown in Formula 1 comprising both a cyclic  
15 carbonate structure and a linear carbonate structure in molecule, when R is methyl group, the compound is 4-carbomethoxymethyl-1,3-dioxolan-2-one; and when R is ethyl group, the compound is 4-carboethoxymethyl-1,3-dioxolan-2-one.

          The compound shown in Formula 1 of the present invention may be  
20 prepared according to the following Scheme 1: a sodium is immediately dissolved by adding in glycerol-1-allyether, then diethyl carbonate is added to obtain 4-allyloxymethyl-1,3-dioxolan-2-one as a first intermediate products, and a palladium carbon and p-toluene sulfonic acid is added to the first intermediate products. Then the mixture is reacted and distilled to obtain 4-  
25 hydroxymethyl-1,3-dioxolan-2-one as a second intermediate product, and the second intermediate product is reacted with methyl chloroformate or ethyl chloroformate, and followed by extracting with methylene chloride solvent to obtain the compound shown in Formula 1.

[Scheme 1]



The compound shown in Formula 1 of the present invention may be used in an electrolyte of lithium ion battery comprising only a lithium salt, or an electrolyte of a cyclic carbonate or a linear carbonate as well as lithium salt. Particular, when the compound is used as a third component in the electrolyte comprising a cyclic carbonate and a linear carbonate in the lithium ion battery including graphitized carbon of high capacity, the lithium ion battery may obtain a high capacity of graphitized carbon, a superior charge-discharge cyclic life and a low temperature performance by removing irreversible capacity.

The electrolyte comprising the compound of the present invention is a non-aqueous solution containing a lithium salt. In particular, the lithium salt is preferably selected from the group consisting of  $\text{LiClO}_4$ ,  $\text{LiCF}_3\text{SO}_3$ ,  $\text{LiPF}_6$ ,  $\text{LiBF}_4$ ,  $\text{LiAsF}_6$ , and  $\text{LiN}(\text{CF}_3\text{SO}_2)_2$ . The electrolyte of the present invention may include ester or carbonate compound which is at least one selected from the group consisting of ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), vinylene carbonate (VC), diethyl carbonate (DEC), dimethyl carbonate (DMC), ethylmethyl carbonate (EMC),  $\gamma$ -butyrolactone, sulfolane, methyl acetate (MA), methyl propionate (MP), and



methyl formate (MF).

The present invention provides the lithium ion battery comprising an anode including high capacity graphitized carbon as an active material, capable of absorbing and releasing lithium ions reversibly, a cathode including lithium-containing transition metal oxide as an active material, 5 capable of absorbing and releasing lithium ions reversibly, a porous separator, and an electrolyte containing a lithium salt and the compound shown in above Formula 1.

To provide the lithium ion battery, the graphitized carbon has preferably a interplanar spacing ( $d_{002}$ ) of less than 0.338 nm in X-ray diffraction of carbonaceous material, and a specific surface area of less than 10  $m^2/g$  measured by Brunauer-Emmett-Teller (BET) method. The lithium-containing transition metal oxide is preferably selected from the group consisting of  $LiCoO_2$ ,  $LiNiO_2$ ,  $LiMn_2O_4$ , and  $LiNi_{1-x}Co_xO_2$  (wherein,  $0 < x \leq 1$ ). 10 In particular, the above battery consists of an anode composed of an active carbonaceous material and polyvinylidene as a binder resin, a cathode composed of lithium-containing transition metal oxide, a conductive agent, and polyvinylidene difluoride as a binder resin. The elements of the battery may be prepared by general method. The lithium ion battery of a high capacity and a superior low temperature performance may be more easily prepared by using the compound shown in Formula 1 of the present invention in the electrolyte. 20

Hereinafter, the present invention is described more in detail through EXAMPLES and COMPARATIVE EXAMPLES. However, the following 25 EXAMPLES are only for the understanding of the present invention, and the present invention is not limited to the following EXAMPLES.

## EXAMPLES

### EXAMPLE 1

(Preparation of 4-carbomethoxymethyl-1,3-dioxolan-2-one)

To 200 g of glycerol-1-allylether (Acros Company), 1.74 g of fresh sodium was added. After sodium was dissociated completely, 196 g of diethyl carbonate was added for 1 hour, and then the reaction mixture was stirred at 130 °C for 12 hrs. After completing the reaction, the mixture was separated into two layers with ethyl ether and distilled water, the combined ethyl ether layer was fractionated to obtain 133g of 4-allyloxymethyl-1,3-dioxolan-2-one as a first intermediate product.

133g of a first intermediate product was added to 1640 ml of methanol, and 19.95 g of 10 wt% of palladium carbon and 13.3 g of p-toluene sulfonic acid were added with stirring under the atmosphere of N<sub>2</sub>, and the mixture was reacted at room temperature for 48 hrs. The resultant reaction mixture was passed through celite 545 to obtain a solution. Vacuum distillation of this solution gave 73 g of 4-hydroxymethyl-1,3-dioxolan-2-one as a second intermediate product.

To 73 g of 4-hydroxymethyl-1,3-dioxolan-2-one was added 63.3 g of pyridine with stirring and 75.6 g of methyl chloroformate was added at 0°C for 1 hr, then the reaction mixture stirred at room temperature for 16 hrs. The reaction solution was passed through a glass filter to remove pyridine HCl salts, and then extracted with methylene chloride and distilled water. Recrystallization was carried out in toluene to obtain 30 g of 4-carbomethoxymethyl-1,3-dioxolan-2-one as a final product.

### EXAMPLE 2

(Preparation of 4-carboethoxymethyl-1,3-dioxolan-2-one)

34g of 4-carboethoxymethyl-1,3-dioxolan-2-one was obtained by the same procedure as in example 1 except that 80g of ethyl chloroformate instead of methyl chloroformate was added to 4-hydroxymethyl-1,3-dioxolan-

2-one as a second intermediate material.

### EXAMPLE 3

(Preparation of an electrolyte)

30 ml of an electrolyte was prepared with mixing in a volume ratio of  
5 EC : 4-carbomethoxymethyl-1,3-dioxolan-2-one (Example 1) : EMC = 41.65 :  
8.35 : 50 and adding 1 M  $\text{LiPF}_6$  solution in glove box. The EC and EMC  
were respectively F-EC and F-EMC, the products of Mitsubishi Chem.  
Company.

### EXAMPLE 4

10 (Preparation of an electrolyte)

30 ml of an electrolyte was prepared with mixing in a volume ratio of  
EC : 4-carboethoxymethyl-1,3-dioxolan-2-one (Example 2): EMC = 41.65 :  
8.35 : 50 and adding 1 M  $\text{LiPF}_6$  solution in glove box. The EC and EMC  
were respectively F-EC and F-EMC, the products of Mitsubishi Chem.  
15 Company.

### COMPARATIVE EXAMPLE 1

(Preparation of an electrolyte)

30 ml of an electrolyte was prepared with mixing in the volume ratio  
of EC : PC : EMC = 41.65 : 8.35 : 50 and adding 1 M  $\text{LiPF}_6$  solution in glove  
20 box. The EC, PC and EMC were respectively F-EC, F-PC and F-EMC, the  
products of Mitsubishi Chem. Company.

### COMPARATIVE EXAMPLE 2

(Preparation of an electrolyte)

30 ml of an electrolyte was prepared with mixing in a volume ratio of  
25 EC : EMC = 50 : 50 and adding 1 M  $\text{LiPF}_6$  solution in glove box. The EC  
and EMC were respectively F-EC and E-EMC, the products of Mitsubishi  
Chem. Company.

### EXAMPLE 5

(Preparation of lithium ion battery)

An anode was prepared by mixing 93 wt% of carbonaceous active material (Osaka gas Company, MCMB-10-28), and 7 wt% of polyvinylidene difluoride (PVDF; Kynar 761 of Elf Atochem Company) in N-methyl-2-pyrrolidinone (NMP) as solvent in mixer (Ika Company) for 2 hrs to obtain an anode slurry, and then by coating the slurry on a copper foil current collector, and followed by drying at 130°C.

A cathode was prepared by mixing 91 wt% of  $\text{LiCoO}_2$ , 3 wt% of PVDF (Kynar 761), 6 wt% of conductive carbon (Lonza Company, KS-6) in NMP in mixer (Ika Company) for 2 hrs to obtain a cathode slurry, and then by coating the slurry on aluminum foil current collector, and followed by drying at 130°C.

18650 cylindrical type of lithium ion batteries were fabricated by winding an anode, a cathode, and a separator (celgard 2400 of Hoechst Celanese Company) between the anode and the cathode, and followed by injecting electrolytes of Comparative Examples 1 and 2, and Examples 3 and 4.

Evaluation of the battery

The charge-discharge experiment was carried out, where the battery was charged to 4.2 V, and then discharged to 3 V at the rate of 0.5 C at room temperature. Then, an experiment of a low temperature performance was carried out, where the battery was left at -20°C for 4 hrs, and then discharged to 2.5 V at the rate of 0.2 C. The results of the charge-discharge experimentals were shown in Table 1.

The battery using the electrolyte of Comparative Example 1 was shown to have good low-temperature performance due to the use of PC, but was shown to have low capacity and a poor charge-discharge cyclic life. Further, the battery using the electrolyte of Comparative Example 2 was shown to have a large capacity, and a poor charge-discharge cyclic life. In

comparison, the battery using the electrolytes of the Example 3 (comprising compound of Example 1), and Example 4 (comprising compound of example 2) was shown to have excellent performance in electric capacity, and the charge-discharge cyclic life, and a low temperature performance.

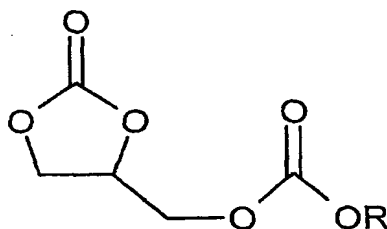
5 [TABLE 1]

	Volume ratio of an electrolyte component (v %)					Evaluation		
	EC	PC	Compound of Example 1	Compound of Example 2	EMC	Initial Capacity (mAh)	Capacity after 200 cyclic (mAh)	Discharge efficiency at -20 °C (%)
Example 3	41.7	0	8.3	0	50	1560	1320	60
Example 4	41.7	0	0	8.3	50	1560	1300	58
Comparative example 1	41.7	8.3	0	0	50	1450	1160	66
Comparative example 2	50	0	0	0	50	1570	1330	10

**WHAT IS CLAIMED IS:**

1. A compound shown in Formula 1:

[Formula 1]



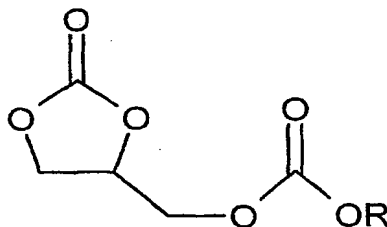
wherein, R is methyl or ethyl group.

2. An electrolyte comprising:

a) a lithium salt; and

b) the following compound shown in Formula 1 comprising both a cyclic carbonate structure and a linear carbonate structure in molecule:

[Formula 1]



wherein, R is methyl or ethyl group.

3. The electrolyte as claimed in claim 2, wherein the electrolyte comprises c) an ester or a carbonate compound which is at least one selected from the group consisting of ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), vinylene carbonate (VC), diethylene carbonate (DEC), dimethylene carbonate (DMC), ethylmethylcarbonate (EMC),  $\gamma$ -butyrolactone, sulforane, methyl acetate (MA), methyl propionate (MP), and methylformate (MF).

4. The electrolyte as claimed in claim 2, wherein the lithium salt is selected from the group consisting of  $\text{LiClO}_4$ ,  $\text{LiCF}_3\text{SO}_3$ ,  $\text{LiPF}_6$ ,  $\text{LiBF}_4$ ,

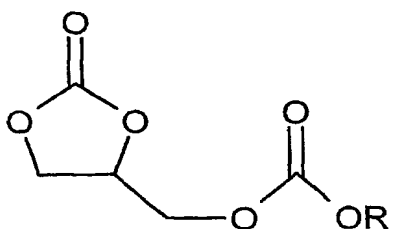
LiAsF<sub>6</sub>, and LiN (CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>.

5. The lithium ion battery comprising an anode including graphitized carbon as an active material, capable of absorbing and releasing lithium ions reversibly, a cathode including lithium-contained transition metal oxide as an active material, capable of absorbing and releasing lithium ions reversibly, a porous separator, and an electrolyte, wherein the electrolyte comprising:

a) a lithium salt; and

b) the following compound shown in Formula 1 comprising both a cyclic carbonate structure and a linear carbonate structure in molecule:

[Formula 1]



wherein, R is methyl or ethyl group.

6. The lithium ion battery as claimed in claim 5, wherein the electrolyte comprises c) an ester or a carbonate compound which is at least one selected from the group consisting of ethylene carbonate (EC), propylene carbonate (PC), butylene carbonate (BC), vinylene carbonate (VC), diethylene carbonate (DEC), dimethylene carbonate (DMC), ethylmethyl carbonate (EMC),  $\gamma$ -butyrolactone, sulforane, methyl acetate (MA), methyl propionate (MP), and methylformate (MF).

7. The lithium ion battery as claimed in claim 5, wherein the lithium salt is selected from the group consisting of LiClO<sub>4</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiPF<sub>6</sub>, LiBF<sub>4</sub>, LiAsF<sub>6</sub>, and LiN (CF<sub>3</sub>SO<sub>2</sub>)<sub>2</sub>.

8. The lithium ion battery as claimed in claim 5, wherein the

graphitized carbon has a interplanar spacing (d002) of less than 0.338 nm in X-ray diffraction of carbonaceous material, and a specific surface area of less than 10 m<sup>2</sup>/g measured by BET method.

9. The lithium ion battery as claimed in claim 5, wherein the lithium-containing transition metal oxide is selected from the group consisting of LiCoO<sub>2</sub>, LiNiO<sub>2</sub>, LiMn<sub>2</sub>O<sub>4</sub>, and LiNi<sub>1-x</sub>Co<sub>x</sub>O<sub>2</sub> (0 < x ≤ 1).



## INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR01/00036

**A. CLASSIFICATION OF SUBJECT MATTER****IPC7 H01M 10/36**

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

IPC7 : H01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

USPAT, FPD, PAJ

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP, A, 05290882 (FUJI ELECTROCHEM CO., LTD.) NOVEMBER 5, 1993 SEE CLAIM1-3	1-2, 5
A	JP, A, 10247519 (SANYO ELECTRIC CO., LTD.) SEPTEMBER 14, 1998 SEE THE WHOLE DOCUMENT	1-9
A	US, A, 5521027 (MATSUSHITA ELECTRIC IND CO., LTD.) MAY12, 1993 SEE THE WHOLE DOCUMENT	3, 6, 9
A	EP, A2, 0398689 (KABUSHIKI KAISHA TOSHIBA, TOSHIBA BATTERY CO., LTD.) NOVEMBER 22, 1990 SEE CLAIM4	4, 7
A	JP, A, 10214638 (HITACHI LTD.) AUGUST 11, 1998 SEE THE WHOLE DOCUMENT	9

☐ Further documents are listed in the continuation of Box C.☐ See patent family annex.

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Metropolitan City 302-701, Republic of Korea

Facsimile No. 82-42-472-7140

Authorized officer

KIM, Jong Jin

Telephone No. 82-42-481-5730

